

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

---

IN RE APPLICATION OF:

Masayoshi HANDA et al.

Application No.: 10/501,507

Confirmation No.: 008270

Filed: July 15, 2004

Art Unit: 1796

For: PROCESS FOR PRODUCING WATER- ABSORBING RESIN EXAMINER: BERNSHTEYN,  
Michael

---

DECLARATION UNDER 37 C.F.R. 1.132

COMMISSIONER FOR PATENTS

P. O. Box 1450

Alexandria, VA 22313-1450

Sir:

I, Hideki YOKOYAMA, residing in Hyogo-ken, Japan, hereby declare  
and state as follows:

1. I am thoroughly familiar with the contents of said U.S. Application  
Serial No. 10/501,507 filed on July 15, 2004, entitled PROCESS FOR  
PRODUCING WATER-ABSORBING RESIN, its prosecution before the United  
States Patent and Trademark Office and the references cited therein.
2. I received a master's degree from Osaka Prefecture University of  
Japan, in the year 1997, majoring in Chemical Engineering.
3. I have been employed in Sumitomo Seika Chemicals Co., Ltd. in  
the year 1997 and have been assigned to the Research Laboratories.

4. I have been involved in the research and development of water-absorbent resin since 1997.

5. The following experiments were conducted by myself or under my direct supervision and control in order to verify that the ranges of the amounts of a reducing agent or an oxidizing reagent as defined in the amended claim 1 has critical significance.

#### EXPERIMENTAL METHODS

The following experiment was conducted under the same conditions as in Example 1 of the present specification (Example C, D and E). By varying the amounts of a reducing agent sodium sulfite, to confirm the range of an amount of a reducing agent that exhibits an effect of discoloration resistance that has critical significance.

#### Example C: Preparation of Sample C

Specifically, 500 ml of n-heptane was added to a 1000-ml five-necked cylindrical round bottomed flask equipped with a stirrer, a reflux condenser, a dropping funnel, a thermometer and a nitrogen gas inlet tube. Thereto was added 0.92 g of sucrose fatty acid ester (surfactant: S-370, manufactured by MITSUBISHI CHEMICAL CORPORATION) having an HLB of 3.0 and dispersed. The temperature of the dispersion was raised to dissolve the surfactant, and thereafter cooled to 55°C.

Separately from the above, 92 g of a 80% by weight aqueous solution of acrylic acid was added to a 500-ml Erlenmeyer flask. Thereto was added

dropwise 102.2 g of a 30% by weight aqueous sodium hydroxide solution while externally cooling, to neutralize 75% by mol of acrylic acid, to give a partially neutralized salt of acrylic acid. Further, 50.2 g of water, 0.11 g of a polymerization initiator potassium persulfate, and 9.2 mg of a crosslinking agent ethylene glycol diglycidyl ether were added thereto, to give an aqueous solution of a monomer for a first step polymerization.

The entire amount of this aqueous solution of the monomer for a first step polymerization was added to the above-mentioned five-necked cylindrical round bottomed flask under stirring and dispersed. After the internal of the system was sufficiently replaced with nitrogen, the temperature of the mixture was raised, and the polymerization reaction was carried out for 1 hour while keeping its bath temperature at 70°C. Thereafter, the polymerization slurry was cooled to room temperature.

Further, 119.1 g of a 80% by weight aqueous solution of acrylic acid was added to a separate 500-ml Erlenmeyer flask. Thereto was added dropwise 132.2 g of a 30% by weight aqueous sodium hydroxide solution while cooling, to neutralize 75% by mol of acrylic acid. Further, 27.4 g of water, 0.14 g of potassium persulfate, and 35.7 mg of ethylene glycol diglycidyl ether were added thereto, to give an aqueous solution of a monomer for a second step polymerization. The aqueous solution was cooled in an ice water bath.

The entire amount of this aqueous solution of the monomer for a second step polymerization was added to the above-mentioned polymerization slurry. After the internal of the system was again sufficiently replaced with nitrogen, the temperature of the mixture was raised, and the second-step polymerization reaction was carried out for 2 hours while keeping its bath temperature at 70°C.

After the termination of the polymerization, 0.42g of a 0.2% by weight aqueous sodium sulfite solution was added to a water-containing gelated product dispersed in n-heptane, and the mixture was stirred for 30 minutes. Thereafter, 5.28 g of a 40% by weight aqueous solution of pentasodium diethylenetriaminepentaacetate was added to a water-containing gelated product dispersed in n-heptane under stirring. Subsequently, water of the water-containing gelated product was removed to the external of the system by azeotropic dehydration. To the resulting gelated product was added 8.44 g of a 2% by weight aqueous solution of ethylene glycol diglycidyl ether, and water and n-heptane were further removed from the mixture by distillation, and the residue was dried, to give 214.1 g of a water-absorbent resin (Sample C).

Example D: Preparation of Sample D

The same procedures as in Example C were carried out except that the amount of the 0.2% by weight aqueous sodium sulfite solution in Example C was changed to 4.22 g, to give 214.5 g of a water-absorbent resin (Sample D).

Example E: Preparation of Sample E

The same procedures as in Example C were carried out except that 16.89 g of a 20% by weight aqueous sodium sulfite solution was added in place of 0.42 g of 0.2% by weight aqueous sodium sulfite solution, to give 217.2 g of a water-absorbent resin (Sample E).

## RESULTS AND DISCUSSION

Yellow Index was measured for each of Sample C, D and E obtained above in the same manner as in the method described in the present specification. Specifically, into a polypropylene vessel having an inner diameter of 3 cm and a depth of 1 cm was evenly placed 2.0 g of a water-absorbent resin. This vessel was allowed to stand for 20 days in a bench-type thermohygrostat set at a temperature of  $50^{\circ} \pm 2^{\circ}\text{C}$  and relative humidity of  $90 \pm 2\% \text{ RH}$ . After allowing to stand, the vessel was taken out from the thermohygrostat, and allowed to stand for some time to cool to room temperature. The entire amount of the water-absorbent resin in the vessel was added to a glass measuring vessel having an inner diameter of 3 cm, and Yellow Index of the water-absorbent resin was determined with a double beam-type spectrophotometer Z-1001DP (manufactured by Nippon Denshoku Kogyo Industries, Co., Ltd.), in which X, Y and Z, tristimulus values of the spectrophotometer are corrected with a standard white board. Yellow Index was calculated by the following equations from X, Y and Z (tristimulus values) of the resulting water-absorbent resin. Similarly, Yellow Index of the water-absorbent resin before the test of allowing to stand in the bench-type thermohygrostat for 20 days was obtained. The above-mentioned determinations were taken thrice, and an averaged value was obtained.

$$\text{Yellow Index} = 100 (1.28X - 1.06Z)/Y$$

Yellow Index for each of these samples are shown in Table I.

Table I

	Sodium Sulfite [%] <sup>*1</sup>	Pentasodium Diethylene- triaminepentaacetate [%] <sup>*1</sup>	Yellow Index	
			After Production	50°C, 90% RH After 20 Days
Ex. 1 of the Present Specification	0.0625	1.25	6.8	7.0
Comp. Ex. 3 of the Present Specification	0	1.25	9.7	13.3
Sample C	0.0005	1.25	10.1	12.5
Sample D	0.005	1.25	9.9	11.4
Sample E	2.0	1.25	2.9	6.3

\*1: amount expressed by weight based on acrylic acid

It can be seen from Table I that:

- 1) when the amount of sodium sulfite is 0.0005% by weight, the water-absorbent resin of Sample C after having allowed to stand for 20 days at 50°C and 90% relative humidity has Yellow Index exceeding 12;
- 2) when the amount of sodium sulfite is 0.005% by weight, the water-absorbent resin of Sample D after having allowed to stand for 20 days at 50°C and 90% relative humidity has Yellow Index of 12 or less; and
- 3) when the amount of sodium sulfite is 2.0% by weight, the water-absorbent resin of Sample E after having allowed to stand for 20 days at 50°C and 90% relative humidity has Yellow Index of 12 or less.

In conclusion, it can be seen i) that from the above 1) and 2), the water-absorbent resin shows a dramatic change in discoloration resistance, as evaluated by Yellow Index, when the amount of a reducing agent or an oxidizing reagent is 0.001 parts by weight, as defined in the amended claim 1, or proximal values thereto; and ii) that from the above 3), the water-absorbent resin has excellent discoloration resistance even when the amount of a reducing agent or an oxidizing reagent is 2.0 parts by weight, an upper limit of the value defined in the amended claim 1.

**Statement Under 18 U.S.C. § 1001**

The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Hideki Yokoyama

Hideki YOKOYAMA

October 31, 2009

Date



**Table A: SUMMARY OF PREVIOUS DECLARATION RESULTS**

Origin of Disclosure	Sample	Reducing Agent	Metal Chelating Agent	Yellow Index	
				After Production	50°C, 90% RH After 20 Days
Present Specification	Ex. 3 of the Present Specification	Added After Polymerization (Gelated) 0.0625% of SS	Added After Polymerization (Gelated) 0.0625% of DTPA-5Na	7.4	8.2
Present Specification	Comp. Ex. 1 of the Present Specification	Not Added	Not Added	10.4	21.1
Present Specification	Comp. Ex.2 of the Present Specification	Added After Polymerization (Gelated) 0.0625% of SS	Not Added	7.7	24.0
Present Specification	Comp. Ex.3 of the Present Specification	Not Added	Added After Polymerization (Gelated) 0.0625% of DTPA-5Na	9.7	13.3
Declaration-1 TABLE	Partially Modified Example of Ex. 3 of the Present Specification	Added Before Polymerization 0.0625% of SS	Added After Polymerization (Gelated) 0.0625% of DTPA-5Na	9.9	14.1
Declaration-2 TABLE I	Partially Modified Example of Ex. 3 of the Present Specification	Added After Drying 0.0625% of SS	Added After Polymerization (Gelated) 0.0625% of DTPA-5Na	11.9	14.2
Declaration-2 TABLE II	Ex.1 of EP' 063 (Hosokawa)	Not Added	Added After Polymerization (Gelated) 0.5% of TPP-Na	12.0	21.2
Declaration-2 TABLE II	Ex.1 of Shimomura	AQUALIC CA + 0.5% of Sodium Thiosulfate	Not Added	10.9	39.5
Declaration-2 TABLE II	Ex.5 of Shimomura	AQUALIC CA + 0.5% of SS + 0.5% of H <sub>2</sub> O <sub>2</sub>	Not Added	11.5	64.1
Declaration-3 TABLE I	Sample A Ex. 1 of EP '063 + Reducing agent	Added After Drying 0.0625%* <sup>1</sup>	Added After Polymerization (Gelated) 0.5% of TPP-Na	11.7	26.3
Declaration-3 TABLE I	Sample B Ex. 1 of EP '063 + Reducing agent	Added After Drying 50%* <sup>2</sup>	Added After Polymerization (Gelated) 0.5% of TPP-Na	10.9	71.4

\*1...by weight based on acrylic acid

\*2...amount of weight based on total weight superabsorbent resin composition.

Abbreviations used in Table A:

Declaration-1 Declaration filed February 16, 2007

Declaration-2 Declaration filed February 04, 2008

Declaration-3 Declaration filed December 31, 2008

SS Sodium Sulfite

DTPA-5Na Pentasodium Diethylenetriaminepentaacetate

TPP-Na Sodium Tripolyphosphate